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Published in:
Applied Physics Letters

DOI:
[10.1063/1.109273](https://doi.org/10.1063/1.109273)

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
1993

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Maior, M. M., Molnar, S. B., Vysochanskii, Y. M., Gurzan, M. I., Loosdrecht, P. H. M. V., Linden, P. J. E. M. V. D., & Kempen, H. V. (1993). New dielectric material for low temperature thermometry in high magnetic fields. *Applied Physics Letters*, 62(21). <https://doi.org/10.1063/1.109273>

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New dielectric material for low temperature thermometry in high magnetic fields

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(Received 3 November 1992; accepted for publication 23 March 1993)

Dielectric experiments on the incommensurate solid solution $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ for $T=1.2$ –200 K reveal a strong temperature dependence of the real part of the dielectric constant for $T<45$ K. The relative dielectric sensitivity $d \ln(\epsilon')/dT \approx 2$ –8 K^{-1} is found to be 2–3 times higher in comparison to widely used glass-ceramic temperature sensors. Moreover, the dielectric constant has a very good time stability and is insensitive to magnetic fields up to 20 T ($dT/dB < 10^{-4}$ K/T). These characteristics make this material a very promising candidate for applications in capacitive temperature sensors for low temperature thermometry in high magnetic fields.

Semiconductor sensors provide some of the most sensitive devices used in thermometry. In the presence of a magnetic field, however, their use is strongly limited due to magnetoresistive effects characteristic for semiconductors. Capacitive sensors using dielectric materials with a strong temperature dependence of the dielectric constant are in general less sensitive, but are relatively insensitive to magnetic fields. At present the commercially available glass-ceramic capacitance thermometers are widely used in thermometry in high magnetic fields.^{1–3} In the present letter we report on a study of a new dielectric crystalline material suitable as a medium for low temperature thermometry in high magnetic fields.

At room temperature $\text{Sn}_2\text{P}_2\text{Se}_6$ has a paraelectric phase. Upon lowering the temperature two phase transitions occur to an incommensurate (IC) ($T_i=220$ K) and a ferroelectric ($T_f=193$ K) phase, respectively. It was recently shown that upon substituting tin atoms in the cation sublattice of $(\text{Pb}_y\text{Sn}_{1-y})_2\text{P}_2\text{Se}_6$ ($y=0$) by isovalent lead atoms the temperature region of existence of the incommensurate IC phase in these materials becomes more extended and simultaneously shifts towards lower temperatures.⁴ For $y < 0.4$ the existence region of the ferroelectric phase has vanished and the IC phase is stable down to 0 K. The dielectric properties of the solid solutions with $y > 0.4$ deviate substantially from those of pure $\text{Sn}_2\text{P}_2\text{Se}_6$. The sudden drop in the dielectric constant, characteristic for the IC-ferroelectric phase transition disappears for $y > 0.4$ (see Fig. 1).

Based on the character of the temperature dependence of the dielectric constant in various representatives of the $(\text{Pb}_y\text{Sn}_{1-y})_2\text{P}_2\text{Se}_6$ solid solution⁵ (see Fig. 1) the $y=0.45$ compound is chosen as the most suitable candidate for further studies. The data shown in Fig. 1 for $y=0.2, 0.4, 0.5, 0.55$, and 0.6 are obtained from sublimation grown crystals. This method provides high quality crystals but yields only small sized crystals, typically a few mm^3 , which limits their practical applicability. In order to obtain larger samples, the $y=0.45$ crystals are grown using the

Bridgman technique. The growing method has been described elsewhere.⁶ The Bridgman grown crystals are generally of somewhat less quality compared to sublimation grown crystals, as is for instance reflected in the lower absolute values of the dielectric constant.

For the experiments platelets perpendicular to the (100) direction are cut ($5 \times 5 \times 1.5$ mm^3) from a $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ crystal. Subsequently Al electrodes are evaporated on the surfaces of the platelets.

Dielectric measurements in the temperature region between 4.2 and 200 K were performed in a dynamical flow cryostat in the quasistatic regime with a cooling/heating rate of 0.5 K/min by means of a General Radio 1615-A transformer bridge operating at 1 kHz, with a measuring field of 4 V/cm. The sample was mounted inside a temperature controlled copper housing. The temperature is controlled by an Oxford DTC 2 temperature controller in combination with a thermocouple mounted in the copper housing. The temperature was measured with a calibrated Allan-Bradley thermoresistor, also mounted in the copper housing. Measurements below 4.2 K have been performed with the copper housing immersed in a pumped liquid He bath cryostat. The temperature was measured using a RuO surface mounted device resistor. For the magnetic field experiments at 1.2 and 4.2 K, the bath cryostat was mounted in a 20 T bitter magnet at the Nijmegen High Field Magnet Laboratory. In this case, the capacitance of the sample was measured using an Andeen Hagerling 2500 A automatic capacitance bridge. A personal computer was used for the data collection.

The temperature dependence of the dielectric constant of $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ shows a smeared maximum at 80 K and a plateau-like behavior with a small thermal hysteresis between 80 and 50 K. The compounds with $y < 0.45$ exhibit the usual hysteresis which is characteristic for the incommensurate phase. For higher Pb concentrations, where the paraelectric-IC phase transition has shifted below 100 K, the thermal hysteresis in ϵ' is absent. For the $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ compound, the region below 50 K is of

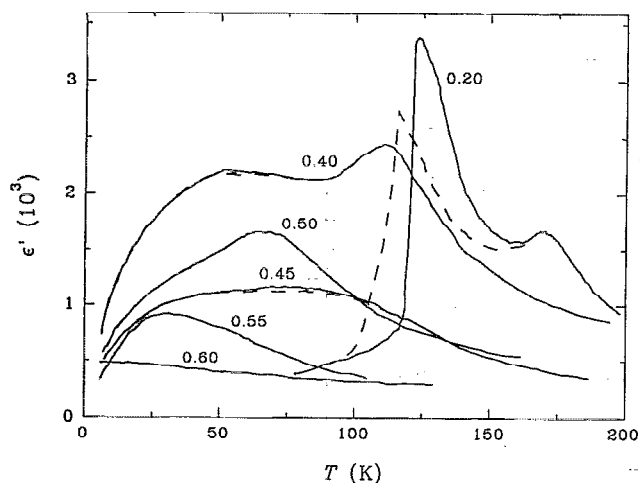


FIG. 1. Temperature dependence of the real part of the dielectric function of the $(\text{Pb}_{0.45}\text{Sn}_{1-y})_2\text{P}_2\text{Se}_6$ solid solutions (solid lines: upon heating, dashed lines: upon cooling) for $y=0.2$; 0.4 ; 0.45 ; 0.5 ; 0.55 ; and 0.6 .

most interest in regard to its possible applicability in temperature sensors. In Fig. 2 we have plotted the dielectric constant and the loss tangent $\text{tg } \delta$ for $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ for temperatures below 50 K. The loss tangent shows a peak near 10 K. The dielectric constant exhibits a frequency dispersion of about $0.5\% \text{ kHz}^{-1}$ in the 1–6 kHz region. Measurements of the time stability of the capacitance and the dielectric loss at 4.2 K showed only very small variations, which could be attributed to pressure variations in the He bath cryostat. The temperature dependence of the absolute $(d\epsilon'/dT)$ and relative $[d \ln(\epsilon')/dT]$ sensitivities of the material as a temperature sensor derived from Fig. 2 are plotted in Fig. 3. The absolute and relative sensitivities reach their maxima at 10 and 7 K, respectively. An important parameter in thermometry, especially at low temperatures, is the amount of heat produced by the used sensor. For capacitance sensors, this heat is produced by dissipation due to dielectric losses. The specific self-heating rate in dielectrics is defined as⁷

$$\frac{\dot{Q}}{V} = \pi f \epsilon' \text{tg } \delta E^2, \quad (1)$$

where V is the sample volume of the dielectric subjected to a time varying electric field E with a frequency f . ϵ' is the

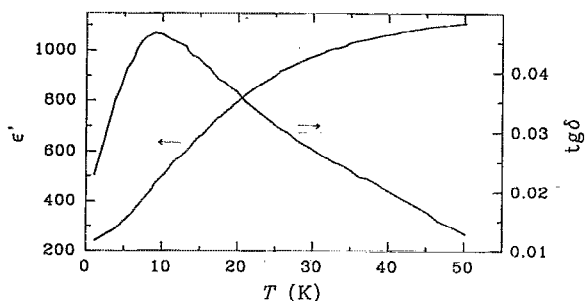


FIG. 2. Temperature dependence of ϵ' and $\text{tg } \delta$ for the $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ solid solutions at $E=4 \text{ V/cm}$ and $f=1 \text{ kHz}$.

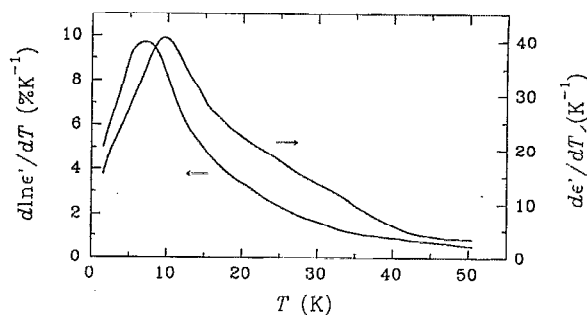


FIG. 3. Temperature dependence of $d\epsilon'/dT$ and $d \ln \epsilon'/dT$ for the $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ solid solution as calculated from Fig. 2.

real part of the dielectric function and $\text{tg } \delta$ the dissipation factor. The calculated temperature dependence of the specific self-heating rate in $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ ($f=1 \text{ kHz}$, $E=4 \text{ V/cm}$) is shown in Fig. 4. It is worth noting that the self-heating rate decreases as a function of temperature below 20 K, i.e., in the temperature region where a low dissipation is of vital importance. Similar to glass-ceramic thermometers, the self-heating rate is very small and lies in the picowatt/cm³ range. The maximum in the self-heating rate corresponds to 40 fW dissipation in the used samples.

Figure 5 shows the magnetic field dependence of the capacitance of $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ for $T=1.2$ and 4.2 K . The measurements plotted here are recorded at a magnetic field sweep rate of 1 T/min . In general it was found that the amplitude of the hysteresis observed in Fig. 5 increases for increasing sweep rates. It is conceivable that part of this hysteresis and its sweep rate dependence is due to local temperature variations in the bath cryostat, caused by for instance eddy currents in the copper heat shields. Similar effects have been observed in dielectric measurements on glass-ceramic materials.¹ The fact that the capacitance does not restore to its initial value is caused by pressure drifts giving rise to a drift in the overall bath temperature. This drift is also observed in the absence of a magnetic field.

In comparing the results on the dielectric parameters of $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ reported here to those of the widely used glass-ceramic thermometers we find that $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ has a significantly higher absolute (2–3 times) and relative (6–7 times) thermal sensitivity.

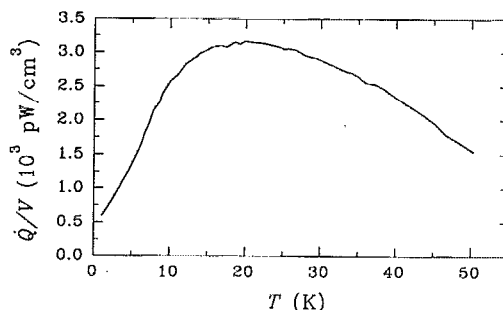


FIG. 4. Specific self-heating rate for the $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ solid solution calculated from Eq. (1).

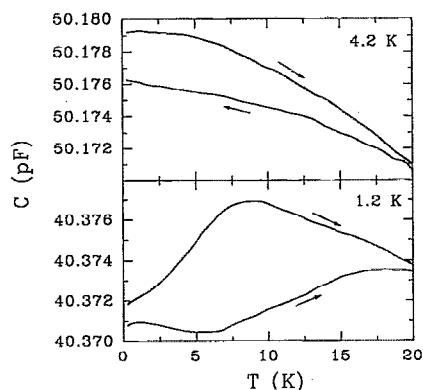


FIG. 5. Effect of the magnetic field on the capacitance of a $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ sample at 4.2 and 1.2 K at a magnetic field sweep rate of 1 T/min.

In particular, we find for the relative sensitivity $d \ln C/dT = 8.5\% \text{ K}^{-1}$, whereas for the glass-ceramic materials a value of $d \ln C/dT = 1.3\% \text{ K}^{-1}$ has been reported.² At 1.2 K the relative sensitivity is about $5\% \text{ K}^{-1}$. Hence, one can expect that even in the sub-Kelvin regime the sensitivity will still be appreciable. This regime is currently under investigation.

The first experiments on the time stability in 30 minutes, as well as on the reproducibility upon thermal cycling show variations in the capacitance corresponding to temperature fluctuations of approximately 5 mK. This value gives an upper bound for the variations, and is determined by the experimental conditions. Further experiments are needed to obtain more accurate data.

The magnetic field dependence of the capacitance corresponds to a field sensitivity of at most 0.1 mK/T for $B < 20$ T. It should be noted, however, that a major part of the field dependence $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ is caused by the experimental conditions, and does not relate to the field dependence of the dielectric parameters.

In respect to the above conclusions, it seems that the $(\text{Pb}_{0.45}\text{Sn}_{0.55})_2\text{P}_2\text{Se}_6$ solid solution provides a most suitable candidate for applications in low temperature thermometry in high magnetic fields.

We are grateful to V. Yu. Slivka for helpful discussions. One of us (M.M.M.) would like to express his thanks to the staff of the Research Institute of Materials of the University of Nijmegen for their hospitality and assistance, and the Dutch and former Soviet Union governments for financial support through a cultural exchange program. Part of this work was financially supported by the Dutch Foundation for Fundamental Research of Matter (FOM).

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